Equation of State Developments in T-1

Nicolas Bock, Leonid Burakovsky, Eric Chisolm, Scott Crockett, Giulia De Lorenzi-Venneri, Tinka Gammel, Denise George, Carl Greeff, J. D. Johnson, Travis Peery, Sven Rudin, and Duane Wallace, T-1

A new unclassified SESAME database was released on June 15, 2005, with five new equations of state (EOS). The update contains four LiH isotopes, one CO_2 modified from an existing EOS, and a material number change for deuterium. A new classified SESAME database was released on June 20, 2005, with two new EOS. Five of the unclassified EOS and one of the classified EOS were produced by T-1; the other classified EOS was produced by Jonathan Boettger, X-1 [1].

A preliminary release of an extension of the PBX 9501 EOS by Sam Shaw, T-14, was given to the user community to test. The extension technique was developed by adapting existing GRIZZLY algorithms.

A new EOS was created for X-4 to meet a special request from a SESAME user. This EOS played a major role in Campaign 4, Major Technical Effort 4.2, Level 2 milestones 1279 and 605 for FY05 and will be officially released next year.

A memo has been written by T-1 and the X-2 Verification & Validation team outlining the SESAME EOS recommended for use in X-2 simulations.

We have continued work on explicit multiphase EOS, in which separate EOS are produced for each phase and the full EOS is constructed by combining the individual phases.

(a) We have incorporated a simple analytic liquid model developed in T-1 into GRIZZLY and OpenSesame. We have validated the model by comparing with analytical work done in T-1 on a two-phase copper EOS [2].

(b) We have produced an EOS for tin that includes the beta and gamma solid phases and the liquid phase and will be included in the next release of the SESAME library [3]. A new treatment of the nuclear contribution, inspired by the Vibration-Transit (V-T) theory of liquid dynamics developed in T-1, was used for the liquid phase, and the phases were combined using an algorithm suggested by a kinetic model used in work on titanium and zirconium. We have also expanded the SESAME format to include new subtables containing the mass fractions of each phase at each grid point, enabling us to map out explicit two-phase regions. The new models and capability to handle the new subtables have been incorporated into OpenSesame.

We are enhancing the reliability of and confidence in the EOS we construct by using not only experimental data and modeling but also density functional theory (DFT) calculations. For example, we are close to completing an improved EOS for beryllium based on extensive calculations that provide a better cold curve and thermal contributions (from calculated phonons). Good agreement between our results and diamond anvil cell data published this year support our new EOS. Similar work on pressure standards (copper, gold, platinum, and tantalum) is providing validation for this approach as well as significant improvements in the accuracy of the EOS of these important materials.

Progress has been made in the development of the V-T theory of liquid dynamics. A new model has been developed to incorporate the effect of transits on the dynamic structure factor $S(q,\omega)$, measured in x-ray and neutron scattering experiments [4].

We consider two distinct contributions to the inelastic scattering due to the two types of motion: independent harmonic vibrations in random valleys and instantaneous transits between valleys. The first contribution, due to vibrational

motion, can be exactly calculated from the random valley Hamiltonian. The second contribution, due to transits, is the object of the model and enters as a correction. By using the model we reproduce, to very high accuracy, $S(q,\omega)$ for liquid sodium, both from molecular dynamics simulations and from recent x-ray inelastic scattering data. Further work to understand the microscopic nature of transits is underway.

Density Functional Theory calculations (GGA+U) of fcc Pu indicate that as the Hubbard on-site Coulomb repulsion U increases, the fcc Pu system goes through a sharp phase transition. Accompanying this transition are sharp drops of the bulk modulus and large values and even negative values of pressure derivatives of the bulk modulus. These effects are consistent with current experimental observations.

Ab initio DFT calculations of the melting of Sn and Na have been performed. The results will be compared with available experimental data and recent constant-energy simulations of superheating of solids to assess the viability of bounding the melting points theoretically (or determining the error bars) of more complex materials at high pressures.

J. D. Johnson and George Baker, T-11, have extended their work on high-temperature expansions of energy to other thermodynamic quantities, such as pressure and entropy [5].

We have continued to develop and evolve the production and maintenance EOS software as the need arises. For example, we have incorporated some more validation checks in the code EOSlibtools. With the help of Jonathan Boettger, X-1, we have also added the capability for machine-independent reading and writing of binary files to accommodate platform-dependent Endian issues. The Ses2d code has also been ported to run on all Unix and Linux platforms.

We have improved the capabilities of the EOS production code OpenSesame in the following ways.

(a) We have replaced the previous command set with a restructured, streamlined set.
(b) We have implemented both the two-phase capability from GRIZZLY and a new multiphase capability.
(c) We have expanded the plotting capabilities by adding a surface plot option (which creates a GMV file) and the ability to plot phase boundaries.
(d) We have standardized the set of units used in inputs to match the units of the SESAME library.
(e) We have expanded the interpolation option to allow any combination of density, temperature, energy, or pressure as long as one of the two supplied variables is either density or temperature.
(f) We have added additional models for the creation of melt and shear tables.
(g) We continue to use version control to maintain a history of source modifications and have updated the documentation and test suite.

The code has also been supplied to several Los Alamos users.

For more information contact Eric Chisolm at echisolm@lanl.gov.

[1] S.D. Crockett, "SESAME Database Release (U)," Los Alamos National Laboratory memorandum t1-2005-06-15-sdc (U), to T-all and X-all (June 15, 2005).
[2] S.D. Crockett, et al., "Testing a liquid EOS model against copper data," to appear in *Shock Compression of Condensed Matter--2005*, M. D. Furnish, Ed. (American Institute of Physics, Melville, New York, 2006).
[3] C.W. Greeff, et al., "SESAME 2161: An explicit multiphase equation of state for tin," Los Alamos National Laboratory report LA-UR-05-9414 (December 2005).
[4] G. De Lorenzi-Venneri and D.C. Wallace, *J. Chem. Phys.* **123**, 244513 (2005).
[5] G.A. Baker, Jr. and J.D. Johnson, *Physica A* **359**, 345 (2006)

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